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INVESTIGATION OF (In, Ga)N/GaN QUANTUM STRUCTURE FOR NITRIDE LASERS

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14. ABSTRACT

Report developed under contract FA8718-08-C-0035. This is the final report for a project to experimentally and theoretically investigate (In,Ga)N/GaN quantum structures for applications in light-emitting devices. The nitride films were grown by molecular beam epitaxy. Their structural properties were characterized by rocking curve x-rays, and their optical quality was assessed by photoluminescence spectroscopy. The main finding is that nitride films grown directly on sapphire substrates were inferior to those grown on template substrates (which are thick bulk-like GaN or AlN layers on sapphire for growth initiation). The lattice constant of GaN cannot be matched to that of most ternary or quaternary films grown on it; compared to sapphire, however, with a mismatch of ~13.5% from GaN, GaN templates are a better alternative substrate. Furthermore, our theoretical analysis suggests that an even better alternative template substrate should be comprised of ternary nitride layers such as In_xGa_{1,x}N or Al_xGa_{1,x}N. unlike sapphire substrates or GaN templates, the lattice constants of the ternaries can be adjusted via their compositions to closely match the average lattice constant of the device layers to be grown on them. This minimizes lattice-mismatch induced strain which has a significant impact on the electronic band structure and on key optical properties of films intended for photonic devices.

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1.0 Executive Summary

This is the final report for a project to experimentally and theoretically investigate indium gallium nitride/gallium nitride ((In,Ga)N/GaN) quantum structures for future applications in light-emitting devices—specifically lasers. The nitride films were grown by molecular beam epitaxy. Their structural properties were characterized by a rocking curve x-ray system, and the optical quality of the films was characterized by use of photoluminescence spectroscopy. The main finding of the project was that nitride films grown on sapphire substrates were of inferior quality compared to those grown on template substrates (which are essentially thick bulk-like GaN or aluminum nitride (AlN) layers grown on sapphire to serve as the base on which to carry out the epitaxial device growth). Even though the lattice constant of GaN cannot be matched to most ternary or quaternary device films grown on the GaN template, the lattice constant of the GaN is not as grossly mismatched from any other nitride films that might be grown on the GaN. The lattice constant of sapphire, on the other hand, is grossly mismatched (about ~13.5% with GaN) from that of most nitride materials. Furthermore, our theoretical analysis suggests that optimal quantum device structures can be grown if the template substrates used are of a ternary nitride type, such as In_xGa_{1-x}N or Al_xGa_{1-x}N. Unlike the sapphire substrates or GaN templates, the composition of the ternaries, and hence their lattice constants, can be adjusted to closely match the average lattice constant of the device layers to be grown on the ternary template. This minimizes the lattice-mismatched induced strain which has a significant impact on the band structure and hence the key optical properties of nitride thin films intended for photonic devices.

The technical results of the project resulted in four journal publications. The list of papers is provided in the report.

2.0 Introduction

This is the final summary report of our work on nitrides carried out from June 2008 to March 2012. The goal of this project was to experimentally and theoretically investigate nitride quantum structures for future applications in photonic device structures.

The nitride compound semiconductor system, generally represented by the quaternary composition as (In,Al,Ga)N, is emerging as an important class of materials for light-emitting and detecting devices. Lasers and light-emitting diodes operating from the ultra-violet (250 nm) to the green (530 - 535 nm) region of the spectrum have been demonstrated. Although these compounds hold much promise for use in the design, growth, and fabrication of devices for a range of applications, much still needs to be done to improve the quality of the beginning materials.

Until recently, the only available substrates for growth of nitride thin films were silicon carbide (SiC) and sapphire. Neither of these substrates is chemically compatible with the nitrides. The only compatibility they share is the crystalline structure. Both have hexagonal crystalline structure, which is similar to that of the nitrides. The major problem however, is that the lattice constants of sapphire and SiC are grossly mismatched from any composition of a nitride film grown on either substrate. To compound this problem, is another that is related to the crystallographic plane of the sapphire on which the nitride films are conventionally grown. Nitride films grown on the c-axis or the (0001) plane of sapphire exhibit polar behavior. This is manifested in the form of an induced electric field that arises due to a polarization charge, and a piezo-electric polarization along the c-axis. Nitride heterostructure films designed for certain devices whose structures are grown on the c-plane must therefore take account of the induced fields. This complicates both the design and the growth of device structures. The piezo-electric polarization, for example, is particular difficult to reliably model because the lattice-mismatch strain on which it depends varies from layer to layer of the device structure.

In recognition of the detrimental effects of foreign substrates used for growth of nitride compounds, two approaches have been undertaken. The first approach has been to grow a thick layer of either an AlN or a GaN film on the sapphire to serve as a template for subsequent nitride films; the second, and perhaps more viable, approach has been to investigate growth bulk GaN substrates. The template approach has lead to major improvements in nitride device performance. Bulk GaN substrates are still not widely available commercially. Only a few companies make them at this point. Most of what they make is for internal consumption (and we have not been able to get any during the course of this work).

Our current view on the growth of thin nitride films is that the new GaN substrate is a step in the right direction. We believe, however, that a holistic approach must still be undertaken. One must consider the entire device structure that is to be grown on the bulk GaN substrate or on the

template substrate during the design process. Although the GaN substrate is chemically compatible with any nitride films grown on top of it, depending on the composition of the films, it is not likely to be latticed-matched to it. A bulk GaN substrate is only lattice-matched to a GaN film on top of it or to an (In,Al)N film of a particular composition.

3.0 Summary of Experimental Work

All the samples investigated during this project were grown by the method of molecular beam epitaxy. Elemental group III sources of Al, Ga, and In were used in their solid metallic form, and a nitrogen plasma source was used for the group V material. The substrate holder was rotated during the growth and the evolution of the films was monitored using a reflection high-energy electron diffraction system (RHEED). The structural quality of the films was characterized by a rocking curve x-ray analysis, and their optical quality was investigated using a photoluminescence spectroscopy system.

In general, we find that the quality of nitride films grown on templates—which are sapphire substrates covered with either thick GaN or AlGaN layers—is better than that of films grown directly on sapphire. The thick template substrates used in our experiments were purchased from a commercial vendor—in our case The main reason why from the company Kyma. sapphire or GaN substrates are not optimal for growth of most nitride films for light emitters or detectors is best illustrated by considering the lattice mismatch problem illustrated in **Figure** 1. The a-axis lattice constant of sapphire a = 4.785 Angstroms (and the c-axis lattice constant

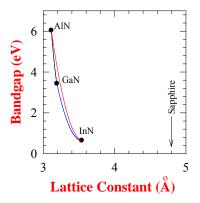


Figure 1: The (a-axis) lattice constant of important binary nitride materials versus the energy band gap. The lattice constant for sapphire is included for reference.

is c = 12.991 Angstroms), while the a-axis lattice constant of GaN is a = 3.186 Angstroms (and the c-axis lattice constant is c = 5.186 Angstroms)[1, 2]. Clearly, the large difference in lattice constants is the main reason why it has been so challenging to grow high quality nitride films on sapphire. Using GaN substrates can improve the situation, but it does not completely remove the lattice-mismatch as any device films grown on any substrate must inevitably include ternary (InGaN, AlGaN, or InAlN) or quaternary (InAlGaN) compositions of the nitrides, each of which has a different lattice constant dependent on the composition. The issue then becomes that of how to manage the strain and stress that result from the inevitable lattice-mismatches. We have devoted some effort to addressing this problem during the course of this project, and discuss briefly some of the considerations in the next section.

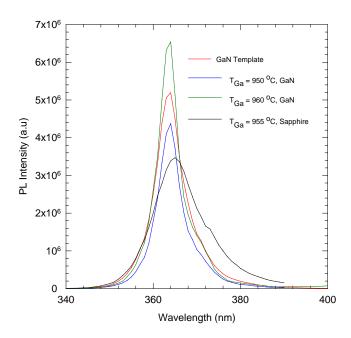


Figure 2: Photoluminescent emission spectra for GaN films grown on sapphire and GaN template substrates for different conditions.

In support of some of the comments made above, we show in Figure 2, the photoluminescence emission spectra from thin (~ 1 μm) GaN films grown on sapphire and on GaN template substrates. As indicated in the figure, the film grown on sapphire (see the inset legend in the figure) shows the broadest emission spectrum; photoluminescent spectral width is an indication of the quality of a film. As the quality improves, the spectrum narrows. This is shown in the other spectra which are for samples grown a GaN template substrate. This trend holds true for other compositions of the nitrides as indicated in Figure 3, where we

show emission from thin ($\sim 1~\mu m$) $Al_{0.72}Ga_{0.28}N$ and GaN films. The difference in intensity for these spectra is attributed to the fact that the $Al_{0.72}Ga_{0.28}N$ film is grown on top of a much thicker GaN under layer. Its quality is therefore much better than that of the GaN that is grown directly on the GaN template on the sapphire.

The key lesson we learned from our experimental work is that the type of the base substrate used for the growth of nitride films is critical to their optical properties. In the absence of optimal substrates, the best choice is a GaN template. If bulk GaN substrates are available, then they would be the first choice. During the course of this project, only Sumitomo, Mitsubishi, and Ammono (in Poland) had reported work on bulk GaN substrates (which not commercially available at the time). We believe however, that a far better choice for the nitride substrates ought to be the ternary nitrides. A ternary substrate would allow specification of a particular lattice constant that would match the application with minimal strain generated by any residual mismatches

4.0 Summary of Theoretical Investigation

Lack of lattice-matching substrates for most nitride light-emitting devices is a serious issue. We investigated this problem extensively during the course of the project. We find that the strain due to lattice-mismatches from unsuitable substrates constraints the emission wavelength of the devices. We summarize in the following, the general approach and main results of our theoretical calculations.

The InGaN alloy is important for photonic applications, particularly green laser device structures. Since all current substrates used for the growth of this alloy have different lattice constants from it, there is always a lattice constant mismatch between an InGaN film and the substrate on which it is grown. It is therefore important to understand the impact of strain on any InGaN/GaN quantum structures used in the active regions of laser devices. Any InGaN/GaN quantum wells grown on either sapphire or native GaN substrates have a certain amount of in-plane biaxial strain because of the difference lattice constants. Arsenideand phosphide-based strained quantum well lasers have been studied for over a decade and it is

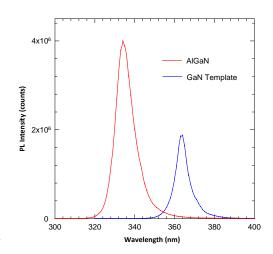


Figure 3: Photoluminescence emission spectra of Al_{0.72}Ga_{0.28}N and GaN thin films grown on GaN template substrates.

widely known that incorporation of strain in the active region of a quantum well laser can have beneficial effects for its optimum performance [3]. In general, the modification of the electronic band structure due to residual strain in compound semiconductors is extremely important since it significantly alters the optical properties of strained films. It is therefore essential to consider the effect of strain on the band structure of a wurtzite InGaN thin film as a function of Indium content, which directly controls the residual strain. We have considered this problem theoretically during the course of this project. Our calculations indicate that confinement effects became smaller (due to reduced conduction band discontinuity) for electrons in the conduction band whereas the opposite trend (due to an increase in valence band discontinuity) holds true for the holes in valence band for a wurtzite-In_xGa_{1-x}N/GaN quantum well with large indium content. However, the relative change seen by holes is much smaller when compared with that for electrons because of the large effective mass of the hole and the smaller change in the valence band discontinuity. Figure 4(a) shows a theoretical plot of the band gap of In_xGa_{1-x}N films grown on GaN substrates as a function of indium mole fraction (x) with and without considering the effect of strain on the electronic band structure. The corresponding values for the residual strain are also shown on the same graph; the strain becomes larger than 3% for indium content exceeding 0.3. A large value of indium mole fraction in the InGaN quantum wells is desirable as this allows the development of green laser diodes on GaN substrates. However, excessive residual strain in the wells degrades the pseudomorhic growth process, leading to a large number of defects or dislocations. In addition to an obvious enhancement in the band gap of the InGaN films with compressive strain, the significant results for light-emitters are shown in Figure 4(b). In Figure 4(b), we plot the energy position of the three valence bands at the Brillouin zone centre (Γ point) as functions of indium mole fraction, i.e., the residual strain. The three valence bands,

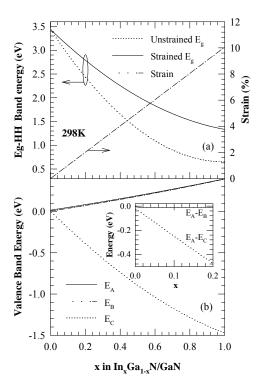


Figure 4(a): The unstrained and strained bad gap of InGaN epilayers versus Indium mole fraction (x) where the corresponding residual strain is shown on the right axis. **Figure 4(b)**: the energy position of three valance bands corresponding to the heavy hole (HH), light hole (LH) and spinorbit crystal-filed split-off hole (SCH) bands labelled as E_A , E_B and E_C respectively. Inset shows the separation between different valance bands for a limited (useful) composition range.

E_A, E_B and E_C correspond to the heavy-hole (hh), light-hole (lh) and the spin-orbit crystal-field split-off hol bands, respectively [4]. The three valence bands are almost degenerate for GaN $(E_A-E_B= -5.4 \text{ meV}, E_A-E_C= -23.5 \text{meV})$; the two lower bands (E_A and E_B) remain more or less degenerate for the entire composition range (E_A- $E_B \le -10$ meV). A small splitting between these two bands can result due to quantumconfinement effects in thin InGaN/GaN quantum wells. The split is caused by the different effective masses of the charge carriers. However, the crystal-field split-off hole band (E_C) is significantly affected by the residual strain and may eventually move out of the potential well in for InGaN/GaN quantum wells for certain values of the residual strain. The separation of E_B and E_C from the lowest valence band (E_A) is plotted in the inset of Figure 4(b); note that the separation between EA from E_C approaches ~ 0.5 eV as soon as the indium mole fraction reaches 0.2. We point out that the degeneracy between the two lowest valence bands can be further lifted by polarization anisotropy in non-polar GaN substrates. This would lead to relatively large gain for light-emitting devices grown on non-polar crystallographic orientations

Theoretical calculations predict that gain in laser structures on m-plane and semi-polar ($11\overline{2}2$) surfaces with 3-nm-thick In_{0.2}Ga_{0.8}N/GaN quantum structures is about three times higher than what is predicted on c-plane surfaces [6,7].

After considering the modification of the electronic band structure due to residual strain, we have calculated the emission wavelength of InGaN/GaN quantum well structures as a function of well thickness and composition. Taking into consideration the band lineup modification by strain in InGaN/GaN quantum wells, we calculate the values of the confinement potentials ΔE_c (ΔE_v) for electrons (holes) in the conduction (valence) band as shown in Figure 5(a). Here ΔE_c (ΔE_v) stands for the discontinuity in the conduction (valence) band for an InGaN/GaN quantum well. The corresponding values for compressive residual strains are also plotted on the same graph. The calculation is restricted to low values of the indium mole fraction (x) in the In_xGa_{1-x}N/GaN quantum well so that the residual strain does not exceed a 3% limit. We arbitrarily chose the 3%

limit for strain from our past work on highly strained InGaAs/GaAs quantum well lasers where devices with reasonable operating characteristics can be made. The limit on strain in either In_xGa_{1-x}As/GaAs or In_xGa_{1-x}N/GaN structures arises from a basic requirement for pseudomorphic growth where one desires the quantum well structures to have any indium mole fraction (for x = 0 to 1), as long as the thickness of the well layer is kept below a socalled critical layer thickness, beyond which the begins develop deleterious structure to dislocations and defects. The critical thickness limit depends on the indium content of the quantum well layer and decreases almost exponentially with x (strain) [3]. Thinner quantum wells can probably accommodate larger indium concentrations, but the drawback is that they then have large blue-shifts due to quantum mechanical effects. Generally, thicker quantum wells grown on c-plane GaN/sapphire tend be inefficient. substrates to compromise is therefore to use thin quantum wells and live with the possibility of carrier overflow. We believe thin quantum well might lead to low efficiency emission.

To assess the impact of carrier overflow in InGaN/GaN quantum-well structures on c-plane substrates, we have numerically solved the Schrödinger equation for a finite, square potential well using the envelope function approximation. Eigen-value solutions of the Schrödinger equation for electrons and holes in

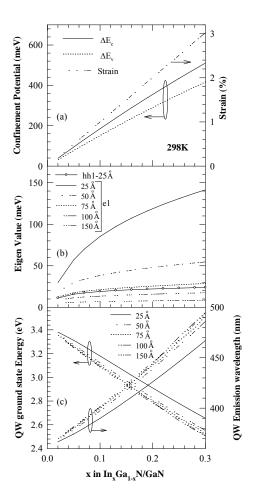


Figure 5(a): The confinement potentials ΔE_c (ΔE_v) for electrons (holes) in the conduction (valence) band respectively, the residual strain is also plotted in the same graph (right axis) for a limited (useful) composition range of InGaN/GaN QW. Figure 5(b): Eigen values e₁ and hh₁ of electrons (holes) in the respective potential wells are plotted as a function of QW composition for different values of QW thickness. Figure 5(c): Energy values corresponding to the ground states transition in InGaN/GaN QWs is plotted along with the corresponding emission wavelength for different values of OW thickness/composition.

the potential well of their respective bands were obtained by solving the resulting transcendental equations. All the materials parameters were taken from the literature and the required parameters for ternary alloys were interpolated by using values from the appropriate binary nitride materials [8]. One of the parameters needed for these calculations is the band offset ratio Q_c . This parameter is defined as $Q_c = \Delta E_c / \Delta E_g$ with ΔE_c being the conduction band offset, and

 ΔE_g the difference between the band gap of the GaN barrier and the strained bulk band gap E_o^s (A) of the InGaN quantum well layer. The values of Q_c can vary widely from 0.38 to 0.83 for wurtzite InGaN/GaN structures [9]. For the present numerical calculations, we have picked $Q_c =$ 55% [10]. The other necessary material parameters for these calculations have been taken from the literature [8]. We plot the energy eigen values of carriers (e₁ for electrons and hh₁ for holes) in an In_xGa_{1-x}N/GaN quantum well in Figure 5(b) as functions of 'x' for different values of quantum well thicknesses. In order to minimize the thermal carrier overflow, it is required that the effective carrier confinement (ΔE_c -e₁ for electrons and ΔE_v -hh₁ for holes) be at least 3kT (~75meV at room temperature), where k is the Boltzmann constant and T the temperature in degrees Kelvin. It can be deduced from Figure 2 that carrier overflow becomes a significant problem for 2.5(5)-nm-thick quantum wells for x < 0.10(0.07), where even the holes may be thermally ejected from a QW for $x \le 0.06$. For InGaN/GaN quantum wells with large indium content (0.2 to 0.3), carrier overflow should be negligible even for thin (2.5 nm) quantum wells. We plot in Figure 2(c), the numerically calculated values of energy for the ground state transition as a function of indium composition in the quantum well for different values of well thicknesses; the exciton binding energy has been taken into account. The corresponding emission wavelengths for the InGaN/GaN quantum well structures are shown on the right hand side of the same graph. It should be noted that the present calculations do not include polarization-induced electric fields which, in general, provide red-shifts of ≤30 nm; the polarization-induced field can also lead to an unwanted reduction in the overlap of the electron and hole wave functions.

If one follows the trend of highly strained InGaAs/GaAs quantum well lasers, where excellent device characteristics are achievable for a residual strain of < 2%, then similar characteristics should be possible for InGaN/GaN quantum well lasers if the indium mole fraction 'x' is chosen below 0.20 and the quantum well thickness remains \leq 3nm for structures on c-plane substrates. For these quantum well parameters, we get an emission wavelength of ≤430 nm as shown in Figure 5(c); this lies in the proximity of the blue portion of the spectrum (when the electric fieldinduced red-shift is taken into account). This is not surprising since we already know that efficient nitride emitters on c-plane GaN substrates cover this portion (violet-blue) of the electromagnetic spectrum. Although the numerical values of several of the materials parameters used in the calculations are not yet well established, the estimate of the emission wavelength is in reasonable agreement with experimental data from the literature [8]. One can now appreciate why the longest possible emission wavelength for nitride quantum well laser diodes on c-plane substrates (within the 3% strain limit) is ~500 nm for a ~3-nm-thick quantum well layer, taking into account the electric field-induced red-shift. This is in good agreement with recent results, where c-plane nitride lasers were shown to operate at 498 nm at room temperature. The emission wavelength can be further red-shifted by about ~20 nm by increasing the thickness of the quantum wells (10-15nm), assuming that the critical-layer thickness is not reached. On the basis of this, our calculations suggest that the longest emission wavelength for nitride lasers on cplane substrates, even with thicker quantum wells would remain below 520 nm. Although these calculations are not valid for m-plane orientations, where additional effects due to polarization anisotropy in the quantum well plane are expected, it is still surprising that the emission wavelengths of nitride lasers on semi-polar/non-polar GaN substrates have not yet exceeded the limit discussed here for c-plane substrates. Because of the absence of spontaneous polarizationinduced fields on non-polar/semi-polar GaN substrates, one ought to be able to grow thicker quantum well layers on these substrates, provided that the critical-layer thickness is not reached. Nevertheless, from Figure 5(c), one sees that the anticipated red-shift for a relatively thick quantum well is probably smaller than the corresponding blue-shift induced by the absence of polarization-induced electric fields. This means that the absence of polarization-induced fields on non-polar/semi-polar GaN substrates may not necessarily guarantee the longest wavelength operation of nitride lasers. In spite of the remarkable results reported on polar, non-polar, and semi-polar GaN substrates so far, it is yet to be determined whether specific crystallographic orientations of GaN substrates would lead to the development of nitride lasers at several wavelengths. Our calculations suggest that the limit on the longest wavelength of nitride laser diodes is constrained by the lattice mismatch rather than by the strength of the polarizationinduced electric field. We believe that it may be possible to develop nitride lasers approaching the green portion of the spectrum (~530- 535 nm) by growing laser structures on non-polar or semi-polar GaN substrates. However, the development of red or infrared nitride lasers appears extremely difficult by simply growing the laser structures on any available crystallographic orientation of the GaN substrate. It is therefore desirable that some new innovative approaches be explored in order to exploit the large band gap range available in the nitride materials.

5.0 List of Journal Publications

- 1. T. K. Sharma and E. Towe, "Impact of Strain on Deep Ultraviolet Nitride Laser and Light-emitting Diodes", Journal of Applied Physics, vol. 109, Issue 8, id. 086104, 2011.
- 2. T. K. Sharma, D. Naveh, and E. Towe, "Strain-Driven Light-polarization Switching in Deep Ultraviolet Nitride Emitters", Physical Review B, vol. 84, id. 035305, 2011.
- 3. T. K. Sharma and E. Towe, "Lattice Constant is Key to Group III-Nitride-Based UV Light Emitters," Laser Focus World, vol. 47, no. 9, pp. 52-54, 2011.
- 4. T. K. Sharma and E. Towe, "On Ternary Nitride Substrates for Visible Semiconductor Light-emitters," Applied Physics Letters 96, id. 191105, 2010.
- T. K. Sharma and E. Towe, "Application-Oriented Nitride Substrates: The Key to Longwavelength Nitride Lasers Beyond 500 nm", Journal of Applied Physics, vol. 107, id. 024516, 2010.

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